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(54) Impact-Resistant Polyolefin Molding Composition

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NOE 92/F 008

Abstract of the disclosure:

Impact-resistant polyolefin molding composition

A polyolefin molding composition essentially comprising 20 to 99% by weight, based on the molding composition, of an isotactic polyolefin which contains no atactic polymer chains, and 1 to 80% by weight, based on the molding composition, of a rubber having a glass transition temperature below -20°C is distinguished by a high impact resistance and hardness, even at low temperatures.

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Description

Impact-resistant polyolefin molding composition

The invention relates to a polyolefin molding composition
5 of an isotactic olefin homopolymer and/or olefin copoly-
mer and a rubber, which has a high hardness and a high
impact resistance even at low temperatures.

Impact-resistant polyolefin molding compositions are
known in principle. They are prepared by mixing, for
10 example, polypropylene and a rubber, such as, for
example, ethylene/propylene rubber (EPN) or ethylene/
propylene/diene rubber (EPDM), in the melt (Angew.
Makromol. Chem. 185/186 (1991) 97; and Polymer, 28 (1987)
47).

15 According to the prior art, the polypropylene used
comprises atactic polypropylene (APP) to the extent of at
least 2 - 10% by weight and polypropylene chains of
widely varying chain length ($M_w/M_n = 5 - 10$) to the extent
of 90 - 98%.

20 Alternatively, a propylene copolymer which also com-
prises, in addition to APP, polypropylene chains and
polymer chains of the comonomer having the build-up
described above, propylene copolymer chains of varying
copolymer contents and likewise varying chain lengths is
25 also used as the polypropylene component.

Moreover so-called reactor blends are known. Those are
composed of APP, polypropylene and polymers of the
comonomer of varying chain length, as well as of
propylene comonomer chains of varying chain length and
30 comonomer contents. They are prepared directly in the
polymerization reactor in one or more polymerization

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steps.

All these molding compositions have the common feature that they have only an unsatisfactory rigidity/hardness at the desired high impact resistance.

- 5 There was thus the object of discovering a molding composition which does not have the adverse properties known from the prior art.

- 10 Surprisingly, it has now been found that if polymers and copolymers which are free from atactic polymer chains (atactic polyolefin; called APO) and have a homogeneous molecular weight distribution (M_w/M_n is preferably less than 5) are used, the abovementioned disadvantages can be avoided.

- 15 The invention thus relates to a polyolefin molding composition essentially comprising 20 to 99% by weight, based on the molding composition, of an isotactic polyolefin which contains no atactic polymer chains and 1 to 99% by weight, based on the molding composition, of a rubber having a glass transition temperature below -20°C .

- 20 The APO-free isotactic polyolefin to be used for the preparation of the polyolefin molding composition according to the invention is a polymer having a narrow molecular weight distribution M_w/M_n of < 5 , preferably < 3.5 , and a uniform chain build-up. Uniform chain build-up is understood as meaning a random distribution of building defects and/or comonomers in the polymer chain.

- 30 APO-free isotactic polyolefins are to be understood as meaning those polyolefins which essentially comprise no atactic polymer chains. These are products (homo- and copolymers) which are prepared using highly stereospecific metallocene/aluminoxane catalyst systems. Corresponding processes are known and are described, for

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example, in EP-A 302,424, EP-A 336,127, EP-A 336,128 and DE-P 40 35 866.0. The isotactic polyolefins prepared by the processes mentioned comprise only very small contents, if any, of atactic polymer chains. As a rule, this content is less than 1% by weight.

In principle, it is also possible for polyolefins which have been prepared using conventional Ziegler catalysts ($\text{MgCl}_2/\text{TiCl}_4/\text{electron donor}/\text{AlEt}_3$) and comprise noticeable APO contents to be converted into polyolefins which are APO-free and therefore according to the invention by extraction of the APO contents with a hydrocarbon. However, because of the associated involved process, i.e. for reasons of cost, such a procedure is not very appropriate.

The isotactic polyolefin to be used according to the invention either is an olefin homopolymer having a molecular weight distribution $M_w/M_n < 5$, preferably < 3.5 , and an isotacticity index (II) of at least 85%, or an olefin copolymer having an M_w/M_n of < 5 , preferably < 3.5 , and an isotacticity index of at least 85%, or the polyolefin is composed of an olefin homopolymer and an olefin copolymer in which the isotacticity index is at least 85% and M_w/M_n of the components is < 5 , preferably < 3.5 .

If the olefin polymer is an olefin homopolymer, it comprises units of an olefin having at least 3 carbon atoms, of the formula $\text{R}^a\text{-CH=CH-R}^b$, in which R^a and R^b are identical or different and are a hydrogen atom or an alkyl radical having 1 to 10, preferably 1 to 6 carbon atoms, or R^a and R^b , with the carbon atoms joining them, form a ring having 4 to 22 carbon atoms. Preferred olefins are propylene, 1-butene, 4-methyl-1-pentene, 1-hexene, pentene and norbornene. Propylene is particularly preferred, i.e. the molding composition according to the invention comprises, in particular, polypropylene.

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If the olefin polymer is an olefin copolymer, it comprises olefin units defined above for the homopolymer and having an isotacticity index of at least 85% to the extent of 99.5 to 50, preferably 99 to 70% by weight, and
5 units of ethylene and/or another olefin of the above definition as the comonomer to the extent of at least 0.5 to 50, preferably 1 to 30% by weight. The comonomer is incorporated randomly with a high regularity. Preferred comonomers are ethylene, 1-butene, 4-methyl-1-pentene,
10 1-hexene, norbornene or pentene. Particularly preferred olefin comonomers comprise propylene units and ethylene comonomer units.

If the polyolefin comprises a mixture of olefin homopolymer and olefin copolymer, the olefin homopolymer has the
15 composition described above. The olefin copolymer comprises an olefin as defined above for the olefin homopolymer to the extent of 20 to 90% by weight, preferably 40 to 90% by weight, and units of ethylene and/or at least one olefin as defined above for an olefin copolymer
20 to the extent of 80 to 10% by weight, preferably 60 to 10% by weight. The comonomer is preferably incorporated randomly. The content of olefin homopolymer in the total polyolefin composition in this case is 20 to 99% by weight, preferably 40 to 95% by weight, and the content
25 of olefin copolymer is 80 to 1% by weight, preferably 60 to 5% by weight.

If it comprises two different polymer components, the polyolefin according to the invention is particularly preferably composed of 40 to 95% by weight of
30 polypropylene (based on the total amount of polyolefin) having an isotacticity index of at least 85%, and 60 to 5% by weight (based on the total amount of polyolefin) of olefin copolymer comprising 40 to 90% by weight of propylene units and 60 to 10% by weight of ethylene units
35 (in each case based on the total amount of olefin copolymer).

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The molding composition according to the invention comprises the isotactic olefin polymer in an amount of 20 to 99% by weight, preferably 40 to 95% by weight.

1 to 80, preferably 5 to 60% by weight of the molding composition according to the invention is a rubber having a glass transition temperature below -20°C . Suitable rubbers are, for example, styrene/butadiene rubbers, silicone rubbers, ethylene/propylene rubbers (EPM) or ethylene/propylene/diene rubbers (EPDM). EPM and EPDM rubbers can additionally also comprise up to 10% of polyethylene, 1,4-hexadiene, norbornadiene or cyclopentadiene can be present in an amount of up to 10% by weight, based on the total amount of rubber, as the diene component.

15 The content of ethylene and propylene is not limited, as long as a glass transition temperature of the amorphous components of less than -20°C is achieved.

20 A typical composition for commercially available EPM rubbers is, for example, 10-60% by weight of propylene units and 90-40% by weight of ethylene units. Of the ethylene units, 0-40% by weight are a pure polyethylene content, and the remainder forms a copolymer content, together with the propylene.

25 EPDM rubbers are of corresponding composition, but 1-10% by weight of a diene of the abovementioned type is also additionally incorporated in the copolymer content, in addition to propylene and ethylene. The melt viscosity of typical EPM and EPDM rubbers is between 0.5 and 300 g/10 minutes (MFI 230/5).

30 The Mooney viscosity (measured at 121°C , ML) is typically between 20 and 80. The tensile stress at 50% elongation is typically 10-300 psi (pounds/square inch, 1 psi = 6894.8 kg/m.second² = 1 Pa).

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Rubbers which can typically be used are on the market, for example, under the currently existing commercial names Vistalon, Exxelor (Exxon Chemicals), Butral (Butral S.A.), Nardel (DuPont) or Buna (Veba).

- 5 In addition to the isotactic olefin polymer and the rubber, the molding composition according to the invention can also contain the customary additives, for example stabilizers, antioxidants, UV absorbers, light protection agents, metal deactivators, free radical scavengers,
- 10 fillers and reinforcing agents, compatibilizing agents, plasticizers, lubricants, emulsifiers, pigments, optical brighteners, flameproofing agents, antistatics and blowing agents.

- The molding composition according to the invention can be
- 15 prepared by the methods customary in plastics processing for mixing polymers and additives.

One possibility is sintering in a high-speed mixer, if all the constituents of the molding composition are pulverulent.

- 20 Another possibility is the use of an extruder having mixing and kneading organs on the screw.

Finally, kneaders such as are employed in the rubber and synthetic rubber industry are also suitable mixing machines.

- 25 The mixing temperature depends on the particular composition of the molding composition and can be determined by simple routine experiments.

- The molding composition according to the invention is distinguished by a high hardness, in combination with
- 30 high impact resistance, in particular even at temperatures below 0°C. In general, this molding composition

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can be used in the form of extruded, injection molded, foamed or blown moldings in all instances where high rigidity, hardness, dimensional stability and scratch resistance in combination with high impact resistance, tear strength and flexural strength are desired.

It can be used in automobile construction, for example, for side lining strips, spoilers, seals, fender linings, bumper materials, truck and tractor fenders, scratch-resistant automobile interior fittings or hub caps.

10 The molding composition according to the invention furthermore is also suitable, for example, for the production of tear-resistant films, membrane filters, fibers and filaments.

15 The following examples are intended to illustrate the invention in more detail.

II = isotacticity index (by ^{13}C -NMR spectroscopy)
 n_{iso} = average isotactic chain length (by ^{13}C -NMR)
 n_{PE} = average polyethylene block length
 20 η_{sp} = viscosity number, measured at 135°C as a 0.1% strength solution in decahydronaphthalene in a capillary viscometer
 $\text{MFI } 230/5$ = melt index at 230°C, 5 kg load, in accordance with DIN 53 735
 25 M_w/M_n = polydispersity (measure of the chain length distribution)
 Determination of the melting points by DSC (20°C/minute)
 BIH = ball indentation hardness (in accordance with DIN 53 456, pressed sheets 4 mm thick)
 30 a_{KV} = notched impact strength according to DIN 53 453, measured on standard small bars (50 x 6 x 4 mm), taken from pressed sheets, with a V notch (flank angle 45°, notch depth 1.3 mm, notch radius 1 mm).

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A ZSK 28 twin-screw extruder (Werner & Pfleiderer) was used for the preparation of the molding compositions.

Example 1

5 A molding composition comprising 90% by weight of isotactic polypropylene, based on the total molding composition, and 10% by weight of a rubber having the following composition was prepared by extrusion:

35.8% by weight of propylene units and 64.2% by weight of ethylene units; 40.4% by weight of the rubber composition was polyethylene and 59.6% by weight was an ethylene/propylene copolymer (EPM) comprising 60.0% by weight of propylene units and 40.0% by weight of ethylene units. The MFI 230/5 of the rubber was 2 g/10 minutes, the viscosity number (VN) was 236 cm³/g, the DSC melting point 15 was 131°C and the DSC glass transition temperature was -56°C. The isotactic polypropylene used had an isotacticity index (II) of 95.0%, an n_{D}^{20} of 49, an MFI 230/5 of 32 g/10 minutes and a melting point of 148°C; further data were: $M_w = 190,000$ g/mol; $M_w/M_n = 2.2$, 20 VN = 174 cm³/g, no APO contents extractable with ether or heptane.

9 kg of the isotactic polypropylene powder were mixed with 1 kg of the rubber, and the mixture was stabilised against chemical degradation under extrusion conditions 25 with 10 g of pentaerythrityl tetrakis[3-(3,5-di-*t*-butyl-4-hydroxyphenyl)propionate]. The temperatures established in the five heating zones of the extruder were 120°C (intake), 150°C, 190°C, 185°C and 180°C (die plate). The extruder screw was operated at 300 revolutions per 30 minute; the melt temperature of the mixture in the extruder was 210°C.

The following data were measured on the molding composition thus produced:

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MFI (230/5) = 31 g/10 minutes; VN = 168 cm³/g;
 BIH = 70 Nmm²; α_{10} = 10.1 mJmm⁻² (23°C);
 5.4 mJmm⁻² (0°C) and 4.3 mJmm⁻² (-20°C).
 Melting point (DSC) = 128 and 148°C, glass transition
 5 temperature Tg = -56°C.

Comparison Example A

Instead of the APO-free (here APP-free; APP = atactic
 polypropylene) isotactic polypropylene of narrow distri-
 bution according to the invention, a polypropylene having
 10 the following data was employed under the same conditions
 as in Example 1:
 MFI (230/5) = 27 g/10 minutes; VN = 220 cm³/g;
 M_w = 312,000 g/mol, M_w/M_n = 8.5; APP content by heptane
 extraction: 2.3% by weight.

15 The molding composition thus obtained had the following
 data:
 MFI (230/5) = 24 g/10 minutes; VN = 245 cm³/g;
 BIH ... Nmm²; α_{10} = ... mJmm⁻².
 Melting point (DSC) = 128/164°C, glass transition
 20 temperature Tg = -56°C.

Examples 2 - 4; Comparison Examples B - D

The procedure was as in Example 1 (Examples 2 - 4) or as
 in Comparison Example A (Comparison Examples B - D), but
 instead of 10% of rubber 15% (Example 2; Comparison
 25 Example B), 25% (Example 3, Comparison Example C) and 40%
 (Example 4, Comparison Example D) of rubber was employed.

The results are summarized in Table 1.

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Table 1

Example	MPI (230/S) [g/10 min]	VN [cm ³ /g]	BIR [Nmm ⁻²]	α_d [mJmm ⁻²]		Melting point (DSC, °C)	Tg (DSC, °C)
				23°C	0°C		
2 VS	27	175	65	13.8	2.1	128/146	-57
	22	235				129/164	-55
3 VC	23	178	54	31.9	19.7	129/138	-58
	19	239				129/162	-57
4 VD	19	179	44	39.1	31.0	130/147	-57
	16	239				129/160	-54

(V = Comparison Example)

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Examples 5 - 8

Examples 1 - 4 were repeated, but a product having the following data was used as the polypropylene according to the invention:

- 5 $\text{TI} = 96.1\%$; $n_{\text{in}} = 53$; $\text{MFI (230/5)} = 4 \text{ g/10 minutes}$; melting point = 151°C ; $M_n = 369,500$, $M_w/M_n = 2.0$; $\text{VN} = 293 \text{ cm}^3/\text{g}$; no AFP contents extractable with ether or heptane.

- 10 This polypropylene was processed by extrusion with 10% by weight (Example 5), 15% by weight (Example 6), 25% by weight (Example 7) and 40% by weight (Example 8) of the rubber to give molding compositions having the data summarized in Table 2.

Comparison Examples E - H

- 15 The procedure was as in Examples 5 to 8, but a polypropylene which is not according to the invention and has the following data was used:
 $\text{MFI (230/5)} = 9 \text{ g/10 minutes}$; $\text{VN} = 302 \text{ cm}^3/\text{g}$;
 $M_n = 288,000 \text{ g/mol}$; $M_w/M_n = 5.7$; AFP by heptane extrac-
 20 tion: 2.7% by weight.

The molding compositions thus obtained, VE (10%), VF (15%), VG (25%) and VH having a 40% rubber content, had the data summarized in Table 2.

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Table 2

Exemple	KPI (230/5) [g/10 min]	VN [cm ² /g]	BIH [Kmm ²]	a _N 23°C	a _N 0°C	-20°C	Melting point (DSC, °C)	T _g (DSC, °C)
5	4	263	67	18.2	7.5	3.5	130/152	-55
VE	8	260	53	13.5	7.0	3.2	129/164	-56
6	5	258	60	26.2	10.7	5.0	129/151	-57
VF	9	257	56	24.2	8.5	3.7	128/152	-55
7	4	254	53	38.5	24.9	15.2	129/151	-56
VG	9	261	48	38.6	23.5	12.7	129/162	-56
8	4	252	43	41.9	40.9	30.5	130/152	-57
VH	7	255	35	41.4	36.3	30.3	128/160	-56

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Example 9

Example 3 was repeated, but a product having the following data was used as the polypropylene according to the invention:

- 5 II = 96.4%, $n_{icn} = 60$; MFI (230/5) = 190 g/10 minutes;
 Melting point = 148°C, $M_v = 154,500$ g/mol, $M_w/M_n = 2.2$;
 VN = 146 cm³/g; no APF contents extractable with ether or heptane.

- The following data were measured on the molding composition prepared by extrusion with the rubber:
- 10 MFI (230/5) = 79 dg/minutes; VN = 170 cm³/g;
 BIH = 72 Nmm⁻²; $\alpha_{kv} = 28.6$ (23°C), 17.5 (0°C) and
 8.7 mJmm⁻² (-20°C).

Example 10

- 15 Example 3 was repeated, but an ethylene/propylene copolymer having the following composition and properties was used as the polyolefin according to the invention:
 Ethylene content 4.3%, incorporation of ethylene with an average ethylene block length of $n_{re} \leq 1.2$, i.e. the
 20 predominant ethylene content is incorporated in isolated units. The II of the propylene sequences was 96.0%.
 MFI (230/5) = 7.0 g/10 minutes; VN 289 cm³/g;
 $M_v = 402,000$ g/mol, $M_w/M_n = 2.0$.
 No APF contents extractable with ether or heptane.

- 25 The following data were measured on the molding composition prepared by extrusion with the rubber:
 MFI (230/5) = 3.5 g/10 min, VN = 272 cm³/g;
 BIH = 50 Nmm⁻²; $\alpha_{kv} = 45.7$ (23°C), 27.9 (0°C) and
 18.4 mJmm⁻² (-20°C).

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Example 11

Example 3 was repeated, but an ethylene/propylene block copolymer which was prepared in two stages and had the following composition and properties was used as the polyolefin according to the invention:

5 12.5% ethylene content; fractionation of the copolymer showed a composition of 76% of polypropylene having an M_w of 96.8k and 24% of an ethylene/propylene copolymer with an ethylene content of 52%, the ethylene being incor-

10 porated both as isolated units and in block form.

$\text{MFI (230/5)} = 4.9 \text{ g/10 minutes}$; $\text{VN} = 326 \text{ cm}^3/\text{g}$;
 $\text{M}_n = 407,000 \text{ g/mol}$, $\text{M}_w/\text{M}_n = 3.1$.
No APP contents extractable with ether or heptane.

The following data were measured on the molding composition prepared by extrusion with the rubber:

15 $\text{MFI (230/5)} = 3.4 \text{ g/10 minutes}$; $\text{VN} = 298 \text{ cm}^3/\text{g}$;
 $\text{BIH} = 39 \text{ Nmm}^{-2}$; u_{kv} : no fractures of the test specimen down to -40°C .

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THE EMBODIMENTS OF THE INVENTION IN WHICH AN EXCLUSIVE
PROPERTY OR PRIVILEGE IS CLAIMED ARE DEFINED AS FOLLOWS:

1. A polyolefin molding composition essentially comprising 20 to 99% by weight, based on the molding composition, of an isotactic polyolefin which contains no
5 atactic polymer chains and 1 to 80% by weight, based on the molding composition, of a rubber having a glass transition temperature below -20°C .
2. A molding composition as claimed in claim 1, wherein the isotactic polyolefin is an olefin homopolymer having
10 an isotacticity index of at least 85%.
3. A molding composition as claimed in claim 1 or 2, wherein the isotactic polyolefin is an olefin homopolymer and comprises units of an olefin having at least 3 carbon atoms, of the formula $\text{R}^a\text{-CH=CH-R}^b$, in which R^a and R^b are
15 identical or different and are a hydrogen atom or $\text{C}_1\text{-C}_{10}$ -alkyl, or R^a and R^b , with the atoms joining them, form a ring having 4 to 22 carbon atoms.
4. A molding composition as claimed in one or more of claims 1 to 3, wherein the isotactic polyolefin is
20 polypropylene.
5. A molding composition as claimed in claim 1, wherein the isotactic polyolefin is an olefin copolymer which comprises units of an olefin having at least 3 carbon atoms, of the formula $\text{R}^a\text{-CH=CH-R}^b$, in which R^a and R^b are
25 identical or different and are a hydrogen atom or $\text{C}_1\text{-C}_{10}$ -alkyl, or R^a and R^b , with the atoms joining them, form a ring having 4 to 22 carbon atoms, and having an isotacticity index of at least 85% to the extent of 99.5 to 50% by weight, and units of ethylene and/or an olefin
30 of the formula $\text{R}^a\text{-CH=CH-R}^b$ to the extent of 0.5 to 50% by weight.

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6. A molding composition as claimed in claim 5, wherein the olefin copolymer is an ethylene/propylene copolymer.

7. A molding composition as claimed in claim 1, wherein the isotactic polyolefin is a mixture of

- 5 a) 20 to 99% by weight of an olefin homopolymer which comprises units of an olefin having at least 3 carbon atoms, of the formula $R^a-CH=CH-R^b$, in which R^a and R^b are identical or different and are a hydrogen atom or C_1-C_{20} -alkyl, or R^a and R^b , with the atoms
10 joining them, form a ring having 4 to 22 carbon atoms, and having an isotacticity index of at least 85%
and
- 15 b) 80 to 1% by weight of an olefin copolymer which comprises units of an olefin having at least 3 carbon atoms, of the formula $R^a-CH=CH-R^b$, in which R^a and R^b are identical or different and are a hydrogen atom or C_1-C_{10} -alkyl, or R^a and R^b , with the atoms
20 joining them, form a ring having 4 to 22 carbon atoms, and having an isotacticity index of at least 85% to the extent of 20 to 90% by weight, and units of ethylene and/or another olefin of the formula $R^a-CH=CH-R^b$ to the extent of 80 to 10% by weight.

- 25 8. A molding composition as claimed in one or more of claims 1 to 7, which additionally comprises stabilizers, antioxidants, UV absorbers, light protection agents, metal deactivators, free radical trapping agents, fillers and reinforcing agents, compatibilizing agents, plasticizers, lubricants, emulsifiers, pigments, optical brighteners,
30 flameproofing agents, antistatics or blowing agents.

9. The use of a molding composition as claimed in one or more of claims 1 to 8 for the production of moldings.

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10. A molding which can be produced from a molding composition as claimed in one or more of claims 1 to 8.

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Abstract of the disclosure:

Impact-resistant polyolefin molding composition

A polyolefin molding composition essentially comprising 20 to 99% by weight, based on the molding composition, of an isotactic polyolefin which contains no atactic polymer chains, and 1 to 80% by weight, based on the molding composition, of a rubber having a glass transition temperature below -20°C is distinguished by a high impact resistance and hardness, even at low temperatures.

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HOR 92/F 008

THE EMBODIMENTS OF THE INVENTION IN WHICH AN EXCLUSIVE
PROPERTY OR PRIVILEGE IS CLAIMED ARE DEFINED AS FOLLOWS:

1. A polyolefin molding composition essentially comprising 20 to 99% by weight, based on the molding composition, of an isotactic polyolefin which contains no
5 atactic polymer chains and 1 to 80% by weight, based on the molding composition, of a rubber having a glass transition temperature below -20°C .
2. A molding composition as claimed in claim 1, wherein the isotactic polyolefin is an olefin homopolymer having
10 an isotacticity index of at least 85%.
3. A molding composition as claimed in claim 1 or 2, wherein the isotactic polyolefin is an olefin homopolymer and comprises units of an olefin having at least 3 carbon atoms, of the formula $\text{R}^a\text{-CH=CH-R}^b$, in which R^a and R^b are
15 identical or different and are a hydrogen atom or $\text{C}_1\text{-C}_{10}$ -alkyl, or R^a and R^b , with the atoms joining them, form a ring having 4 to 22 carbon atoms.
4. A molding composition as claimed in one or more of claims 1 to 3, wherein the isotactic polyolefin is
20 polypropylene.
5. A molding composition as claimed in claim 1, wherein the isotactic polyolefin is an olefin copolymer which comprises units of an olefin having at least 3 carbon atoms, of the formula $\text{R}^a\text{-CH=CH-R}^b$, in which R^a and R^b are
25 identical or different and are a hydrogen atom or $\text{C}_1\text{-C}_{10}$ -alkyl, or R^a and R^b , with the atoms joining them, form a ring having 4 to 22 carbon atoms, and having an isotacticity index of at least 85% to the extent of 99.5 to 50% by weight, and units of ethylene and/or an olefin
30 of the formula $\text{R}^c\text{-CH=CH-R}^d$ to the extent of 0.5 to 50% by weight.

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